

LA-UR-17-27132

Approved for public release; distribution is unlimited.

Title: Isotopic Ratios of Samarium by TIMS for Nuclear Forensic Application

Author(s): Louis Jean, James
Inglis, Jeremy David

Intended for: Keepin Program NSSC Summer Program Debrief

Issued: 2017-08-08

Disclaimer:

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. By approving this article, the publisher recognizes that the U.S. Government retains nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

Isotopic Ratios of Samarium by TIMS for Nuclear Forensic Application

James Louis-Jean^{1, 2}, Jeremy D. Inglis¹

¹Los Alamos National Laboratory

C-NR: Nuclear & Radiochemistry

Mass Spectrometry & Clean Chemistry Lab

TA-48,0045

²University of Nevada-Las Vegas

Department of Chemistry,

Radiochemistry program



LA-UR-XXXXXX



Operated by Los Alamos National Security, LLC for the U.S. Department of Energy's NNSA

James Louis-Jean (C-NR, TA-48,0045)



- **Educational Background**

- BA, Florida Memorial University, 2014
- PhD, 3rd year, University of Nevada-Las Vegas, 2015-present

- ***Research Group/Mentor***

- *C-NR: Nuclear & Radiochemistry*
 - *Mass Spectrometry & Clean Chemistry Lab*
 - Jeremy D. Inglis, PhD



James Louis-Jean



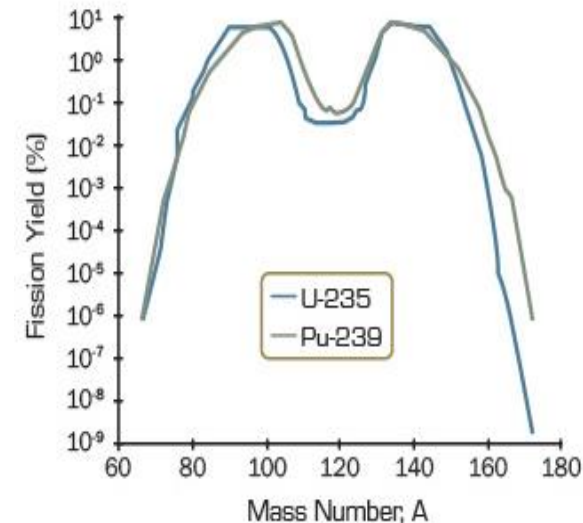
Dr. Jeremy Inglis

- **Research Topics**

- Summer Research Topic: Isotopic Ratios of Samarium by TIMS for Nuclear Forensic Application
- Thesis Topic: Fundamental Chemistry of ^{99}Tc for Nuclear Waste Application

Research Overview and Motivation

- Nuclear forensics: evidence of nuclear materials (NMs), trafficking, enrichment, and signatures.
- Various form of material can be recovered for evidence: dust and radioactive debris.
- In nuclear devices, the neutron induced fission of ^{235}U and ^{239}Pu produce various rare earth elements (REEs).
- Nd, Sm, and Gd are produced in measurable quantities in non-natural isotopic abundances.
- They have many stable isotopes with high thermal neutron cross sections ($>1000\text{b}$).
- Results in depletion of isotopes with large cross sections (^{149}Sm) and enrichment of isotopes of those with low cross sections; consequently proportional to the neutron fluence.



Research Overview and Motivation

- The isotopic ratio of Nd, Sm, and Gd can provide important information regarding fissile material (nuclear devices, reactors), neutron environment, and device yield.
- These studies require precise measurement of Sm isotope ratios, by either TIMS or MC-ICP-MS.
- There has been an increasing trend to measure smaller and smaller quantities of Sm bearing samples.
- In nuclear forensics 10-100 ng of Sm are needed for precise measurement.

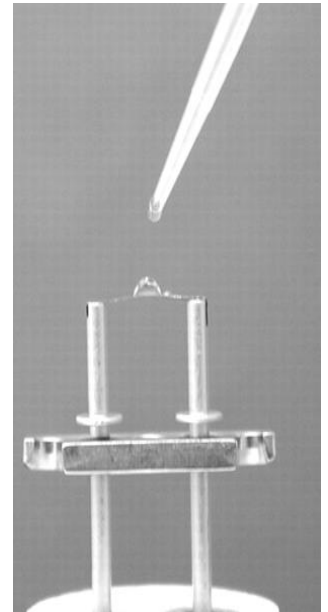
Goal

→ To measure sub-ng Sm samples using TIMS for nuclear forensic analysis.

| | | | | | | | | | | | | | |
|---------------------------|---------------------------|--------------------------------------|---------------------------------------|-------------------------------------|-----------------------------|----------------------------|----------------------------|----------------------------|----------------------------|----------------------------|----------------------------|----------------------------|---------------------------|
| ¹⁴⁴ Sm 3.07 | ¹⁴⁵ Sm 340d | ¹⁴⁶ Sm 1.0e8a | ¹⁴⁷ Sm 14.99 1.2e11a | ¹⁴⁸ Sm 11.24 7e15a | ¹⁴⁹ Sm 13.82 | ¹⁵⁰ Sm 7.38 | ¹⁵¹ Sm 90a | ¹⁵² Sm 26.75 | ¹⁵³ Sm 1.9d | ¹⁵⁴ Sm 22.75 | | | |
| | ¹⁴⁴ Pm 360d | ¹⁴⁵ Pm 17.7a | ¹⁴⁶ Pm 5.53a | ¹⁴⁷ Pm 2.82a | ¹⁴⁸ Pm 5.37d | ¹⁴⁹ Pm 2.21d | ¹⁵⁰ Pm 2.68h | ¹⁵¹ Pm 1.18d | ¹⁵² Pm 4.1m | ¹⁵³ Pm 5.4m | ¹⁵⁴ Pm 2.7 m | | |
| | | ¹⁴⁴ Nd 23.8 2.4e15a | ¹⁴⁵ Nd 8.3 | ¹⁴⁶ Nd 17.2 | ¹⁴⁷ Nd 10.98d | ¹⁴⁸ Nd 5.7 | ¹⁴⁹ Nd 1.73h | ¹⁵⁰ Nd 5.6 | ¹⁵¹ Nd 12.4m | ¹⁵² Nd 11.4m | ¹⁵³ Nd 32s | ¹⁵⁴ Nd 25.9s | |
| | | | ¹⁴⁴ Pr 17.3m | ¹⁴⁵ Pr 5.98h | ¹⁴⁶ Pr 24.2m | ¹⁴⁷ Pr 13.4m | ¹⁴⁸ Pr 2.27m | ¹⁴⁹ Pr 2.3m | ¹⁵⁰ Pr 6.2s | ¹⁵¹ Pr 18.9s | ¹⁵² Pr 3.2s | ¹⁵³ Pr 4.3s | ¹⁵⁴ Pr 2.3s |

Research Approach

- **Different recipes for Sm ions emission**
 - H₂O:Pt/Re solution
 - 5-1; 10-1; 20-1; 30-1
 - Samarium standard (2 ng/μL)
 - H₃PO₄ (0.1 N): 1 μL
- **Samples Prep:**
 - Samples were loaded on degassed Re filaments:
 - (1st: H₃PO₄; 2nd: Pt/Re; 3rd: Sm std)
- **Measurements:**
 - Samples were measured on a GV IsoprobeT in static mode using 7 Faraday collector cups.



Summary of Results

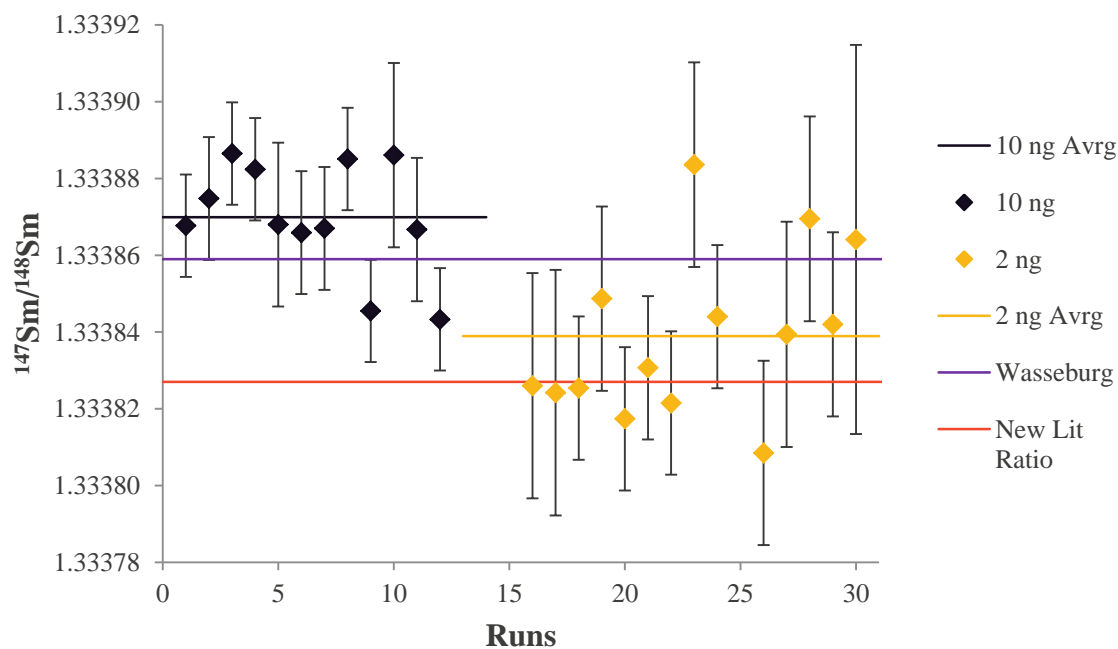
| | *2 ng Sm Std Pt/Re | | | | 10 ng Sm Std |
|----------------|-----------------------|-----------|-----------|-----------|--------------|
| | 5-1 | 10-1 | 20-1 | 30-1 | TaF gel |
| Average | 1.333845 | 1.3338352 | 1.3338456 | 1.3338381 | 1.333869917 |
| 2std | 5.47E-05 | 3.21E-05 | 4.45E-05 | 4.51E-05 | 2.88905E-05 |
| ppm | 41.042351 | 24.101456 | 33.397234 | 33.789168 | 21.65915356 |
| Wasserburg | 1.333859 | | | | |
| New Lit. Ratio | 1.333827 | | | | |

Samples loaded with the Pt/Re 10-1 solution showed better result.

- Load size: 2ng Sm std
- $^{147}\text{Sm}/^{148}\text{Sm}$ isotopic ratio

Different load sizes and recipes

- **2 ng Sm std with Pt/Re (10:1)**
 - Show better agreement with the new literature reported ratios (newly accepted).
- **10 ng Sm std with TaF**
 - Show better agreement with Wasserburg (previously accepted ratio).



Conclusion

- **Measured sub-ng of Sm by TIMS.**
 - 2 ng of Sm std
- **Investigated an effort to make precise Sm isotopic measurements.**
 - Pt/Re loading recipes
- **Improved the amount of materials which can be analyze for forensic.**
 - Trinitite: 100 ng Sm/ g
- **Possibility to determine the number of fissions by measuring stable isotopes of Sm.**
 - Samrium: $^{144}, ^{147}, ^{148}, ^{149}, ^{150}, ^{152}, ^{154}\text{Sm}$
 - ^{149}Sm : high neutron cross section ($\sim 40,400$ barns at 0.025eV)
 - $^{149}\text{Sm}(n,\gamma)^{150}\text{Sm}$

FUTURE DIRECTIONS

- **Continue to investigate the Pt/Re 5:1 recipes.**
- **Study and compare other recipes**
 - TaF/H₃PO₄; TaF/Pt; Pt/Re /H₃PO₄

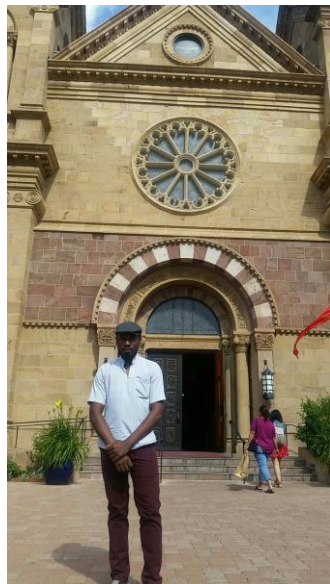
Acknowledgments & Summer Fun

Acknowledgments

- Los Alamos National Laboratory; *C-NR: Nuclear & Radiochemistry; Mass Spectrometry & Clean Chemistry Lab*
- Dr. Jeremy D. Inglis
- Nuclear Science & Security Consortium, *NSSC*



Santa Fe exploration.
Picture by: Maria Pinilla



Santa Fe exploration.
Picture by: Maria Pinilla



Backyard deer watching after work.



Fourth of July celebration